

Analysis of accumulation factor of ^{137}Cs in bottom sediments of lakes and rivers with concentration of the radionuclide close to background levels

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Abstract. The two-chamber (water and bottom sediments) model was used to analyze factors influencing magnitude of ^{137}Cs accumulation factor in bottom sediments of rivers and lakes with concentration of the radionuclide close to background levels. The most probable values of the factor are given. It is proposed to use the factor for assessments of ^{137}Cs activity in water on the base of its observed activity in bottom sediments.

One of the most important radioactive substances in water radioecology is ^{137}Cs . Its presence in surface water-bodies can result from present-day operation of plants of nuclear fuel cycle as well as from fallouts related to past nuclear accidents and atmospheric nuclear weapons testing. Present-day background activity of ^{137}Cs in water of rivers and lakes of Russia does not exceed 10 Bq/m^3 . But even such small activity can produce doses up to 0.016 mSv/year . This exceeds the commonly used in Russia dose limit for normally operated nuclear power plants (NPP) (0.01 mSv/year). That's why a representative assessment of concentration of ^{137}Cs in components of water-bodies is necessary in some cases to separate contribution of global fallouts from contribution of present day NPP's.

If concentration of ^{137}Cs is close to background, its accurate measurement can be quite laborious. First of all it is due to necessity to concentrate the radioactive substance from large samples (200 – 600 liters). The necessary size of samples depends on activity of ^{137}Cs per unit volume and on sensitivity of spectrometric detectors. In the same time bottom sediments accumulate ^{137}Cs . Thus necessary size of a sample of bottom sediments usually does not exceed 1 kg.

That's why observations of specific activity of ^{137}Cs in water are often scarce and not representative in contrast to mass observations of specific activity of ^{137}Cs in bottom sediments.

In number of cases it is necessary only to do preliminary estimations of possible specific activity of ^{137}Cs in water. For this purpose, the possibility to use numerous observations of specific activity in bottom sediments for assessments of activity in water can be very useful. The assessments can be carried out with the use of ^{137}Cs accumulation factor in bottom sediments.

The factor is steady state ratio of specific activities of ^{137}Cs in water and in bottom sediments.

$$K = \frac{C_b}{C_w},$$

(1)

where, ' C_b ' is the activity of ^{137}Cs in bottom sediments per unit mass (air-dry weight), Bq/kg; ' C_w ' is the activity of ^{137}Cs in water per unit volume, Bq/m³ [3].

The accumulation factor depends on many factors. Moreover, the accumulation factor usually is not constant, unless the transport of the radioactivity is steady.

In the work the two-chamber (water and bottom sediments) model [4] was used to analyze factors influencing magnitude of the accumulation factor in rivers and lakes with concentration of ^{137}Cs close to background.

The model takes into account radioactive decay, sorption and desorption of radioactive substances by suspended particles and bottom sediments, sedimentation and resuspension of contaminated particles, diffusion of radionuclides between water and upper layer of bottom sediments, outflow of radioactivity due to water flow, technological water losses, filtration of water through bottom and walls of a water object.

The system of equations describing the processes in water and upper layer of bottom sediments of a lake is as follows:

$$\begin{cases} \frac{\partial C_w}{\partial t} = -\lambda C_w - \frac{C_w \nu \alpha_{Tw}}{H} + \frac{C_b \psi \alpha_{Tb}}{H} + \frac{\beta}{H} (\alpha_{Pb} C_b - \alpha_{Pw} C_w) - \frac{Q}{V} C_w - \frac{Q_f}{V} C_w \alpha_{Pw} + F \\ \frac{\partial C_b}{\partial t} = -\lambda C_b + \frac{C_w \nu \alpha_{Tw}}{h} - \frac{\psi C_b \alpha_{Tb}}{h} - \frac{\beta}{h} (\alpha_{Pb} C_b - \alpha_{Pw} C_w) - \frac{W_c C_b \alpha_{Tb}}{h} - \frac{\gamma \alpha_{Pb} C_b}{h} \end{cases} \quad (2)$$

Where: ' t ' is time, s; ' C_w ' and ' C_b ' are the activities of ^{137}Cs per unit volume of water and bottom sediments respectively, Bq/m³; ' λ ' is the radioactive decay constant, s⁻¹; ' H ' is the average depth, m; ' V ' is the volume of a lake or of its part, m³; ' Q ' is the flowage of a lake, m³/s; ' Q_f ' is the intensity of water filtration through bottom and walls, m³/s; ' F ' describes sources of ^{137}Cs , Bq/(m³*s); ' h ' is the thickness of the upper (effective) layer of bottom sediments, m; ' β ' is the coefficient of diffusive mass exchange of ^{137}Cs between water column and upper layer bottom sediments, m/s; ' γ ' is the coefficient of diffusive mass exchange of ^{137}Cs between upper and lower layers of bottom sediments, m/s; ' ν ' is the effective sedimentation rate of suspended particles, m/s; ' ψ ' is the intensity of resuspension, m/s; ' W_c ' is the silting rate, m/s; ' α_{pw} ' and ' α_{pb} ' are fractions of ^{137}Cs dissolved in water column and in upper layer of bottom sediments respectively, dimensionless; ' α_{Tw} ' and ' α_{Tb} ' are fractions of ^{137}Cs sorbed on particles in water column and in upper layer of bottom sediments respectively, dimensionless.

Intensity of resuspension can be assessed on the base of balance of suspended particles in water:

$$\psi = \nu \frac{S_1}{m} - W_c,$$

Where ' S_1 ' is the concentration of suspended particles in water, kg/m³; ' m ' is the air-dry weight of unit volume of bottom sediments, kg/m³.

Fractions of a radionuclide sorbed on particles and in solute can be determined with the use of well-known dependencies [5]:

$$\alpha_{Pw} = \frac{1}{1 + S_1 k_{dw}} \quad \alpha_{Tw} = \frac{S_1 k_{dw}}{1 + S_1 k_{dw}} \quad \alpha_{pb} = \frac{1}{1 + m k_{db}} \quad \alpha_{tb} = \frac{m k_{db}}{1 + m k_{db}}$$

where ' k_{dw} ' is the partitioning coefficient of a radionuclide between water and suspended matter, m³/kg; ' k_{db} ' - the partitioning coefficient of a radionuclide between pore water and solid phase, m³/kg.

For steady-state problems, time derivatives in (2) are equal to zero and the accumulation factor in bottom sediments can be determined with the use of expression:

$$K = \frac{(\beta + \nu S_1 k_{dw}) * k_{db}}{(\lambda h (1 + m k_{db}) + \nu S_1 k_{db} + \beta + \gamma) * (1 + S_1 k_{dw})} \quad (3)$$

Similar two-chamber model of a river takes into account processes of advection and turbulent dispersion in addition to the processes mentioned above [4]. It is easy to show that for the river model the accumulation factor can be also determined with the use of the expression (3).

One can see in (3) that there is no direct dependence of ' K ' from depth, sedimentation rate and constituents of water budget (intensity of water outflow, filtration, evaporation). Nevertheless, it is necessary to keep in mind that the accumulation factor can depend on these parameters indirectly. For example depth and sedimentation rate influence the coefficient of diffusive mass-exchange between water and bed sediments [4].

Let us carry out a brief comparative analysis of influence of the expression (3) parameters on magnitude of the ¹³⁷Cs accumulation factor. In Table 1 one can find range of variation and typical value for every parameter in (12). The typical values are for water objects with bed sediments of silt or silty sand. Presence of silt in bed sediments indicates indirectly on influence of sedimentation on exchange of radioactivity between water and bed sediments.

If pebble or sand prevails in bed sediments, influence of sedimentation is usually negligible (erosion prevails). Thus accumulation factor there is determined by sorption and diffusion and is usually minimal.

Let us discuss assessment of the accumulation factor for a water object with typical value of the parameters (see Table 1).

Table 1. Parameters influencing migration of ¹³⁷Cs between water column and bottom sediments [4,6].

Parameter	Designation	Unit measure	Used value	Typical value
Concentration of suspended particles in water	S_1	g/m ³	30	5 ÷ 150
Air-dry weight of unit volume of bottom sediments	m	kg/m ³	1000	500 ÷ 2000
Sedimentation rate of				

suspended particles	ν	m/s	$5 \cdot 10^{-5}$	$3 \cdot 10^{-5} \div 8 \cdot 10^{-5}$
Thickness of the upper layer of bottom sediments	h	m	0.1	$0.01 \div 0.2$
Diffusive mass exchange coefficient	β	m/year	0.6	$0.2 \div 5.0$
Partitioning coefficient of ^{137}Cs between water and suspended matter	k_{dw}	m^3/kg	27	$5 \div 500$
Partitioning coefficient of ^{137}Cs between pore water and solid phase	k_{db}	m^3/kg	5	$0.5 \div 50$

For assumed values of parameters the accumulation factor in bottom sediments is $14.2 \text{ m}^3/\text{kg}$.

The analysis shows that 'h' doesn't influence the factor significantly. For silty bottom sediments and any values of 'm', a change of 'h' in 10 times leads only to 10% change of 'K'. It is very important, because setting of thickness of the upper layer of bottom sediments is rather voluntary. If there was significant dependence of 'K' from that parameter, it would lead to unreliability of assessment results. Influence of 'm' and ' β ' on the accumulation factor is also insignificant and does not exceed 3.5%. The sedimentation rate ' ν ' influence 'K' noticeably (up to 25%) only if ' S_1 ' is less than 10 g/m^3 .

The analysis of parameters in (3) shows, that 'K' depends mostly on two parameters – the partitioning factor ' k_{dw} ' and the concentration of suspended matter in water ' S_1 '. These two parameters determine transfer of ^{137}Cs between water and bottom sediments due to sedimentation and resuspension of contaminated particles. Decrease of ' k_{dw} ' from 27 to $10 \text{ m}^3/\text{kg}$ leads to decrease of 'K' almost in 2 times, while increase of ' k_{dw} ' from 27 to $50 \text{ m}^3/\text{kg}$ leads to increase of the accumulation factor by 34%. Concentration of suspended matter in water ' S_1 ' significantly influences specific activity of ^{137}Cs in water and consequently it influences the accumulation factor. An increase of ' S_1 ' leads to decrease of 'K'.

Concentration of suspended matter in water objects significantly depends on erosion of soils on its watershed, prevalent type of bottom sediments, depth and carrying capacity of the flow. The carrying capacity is influenced by seasonal variations and hydrometeorological conditions. In rivers the carrying capacity is determined predominantly by flow velocity. On rapids it is higher and erosion of bed sediments is possible. On reaches it decreases and that leads to sedimentation of some part of suspended particles.

Concentration of suspended matter varies significantly from season to season. In rivers it is maximum during floods and minimum in summer and in winter.

In lakes and ponds the variation of suspended matter concentration in spring and in summer can be caused by resuspension due to wind and waves. The resuspension caused by wind and waves depends on quantity of plankton. In shallow ponds concentration of suspended matter in spring is usually between $30 \div 70 \text{ g/m}^3$. In summer it reaches $70 \div 130 \text{ g/m}^3$ due to development of plankton.

Unlike other parameters in (3), concentration of suspended matter can be easily measured with the use of methods of filtration or using electronic devices.

It is convenient to use observed values of 'S₁' to adjust assessment of the accumulation factor of ¹³⁷Cs in bottom sediments, using linear dependence:

$$K_a = a \cdot K, \quad (4)$$

Where 'K_a' is the adjusted to real conditions assessed value of the accumulation factor, m³/kg; 'K' is *the* average, calculated under typical values of migration parameters, accumulation factor of ¹³⁷Cs equal to 14.2 m³/kg; a - the adjustment coefficient (see Table 2), dimensionless.

For average annual 'S₁' equal to 5 g/m³ adjusted assessed value of the accumulation factor 'K_a' is 18.5 m³/kg. Increase of average annual 'S₁' to 200 g/m³ leads to decrease of 'K_a' to 4.1 m³/kg.

Table 2. Values of the adjustment coefficient 'a' depending on concentration of suspended matter 'S₁'.

Concentration of suspended matter in water 'S ₁ ', g/m ³												
5	10	20	30	40	50	60	70	80	90	100	150	200
Adjustment coefficient 'a', dimensionless												
1.30	1.30	1.15	1.00	0.88	0.78	0.71	0.64	0.58	0.53	0.48	0.37	0.29

In table 3 one can see comparison of observed activities of ¹³⁷Cs in rivers of Moscow region [7, 8] and assessments carried out with the use of the accumulation factor. The observed and assessed values of activity of ¹³⁷Cs per unit volume are in a good agreement. The authors do not intend to use the examples to prove the reliability of the assessment scheme. In real life difference between observed and assessed values can be significant.

Table 3. Comparison of observed and assessed activities of ¹³⁷Cs in water.

Water object	Bottom sediments (Silty), Bq/kg (air-dry weight)	Water, Bq/m ³	
		Observation	Assessed value
Volga River	8	0.9	0.9
The Moscow Canal	7	1.0	0.8
Moscow River	10	1.7	1.1
Moscow River Moscow Region outside Moscow city)	5	0.9	0.5
Oka River	14	1.0	1.5
Average	8.8 ± 2.6	1.1±0.24	0.96±0.23

CONCLUSION

Indeed, the proposed model is simple and due to taken conservative assumptions can not be completely adequate in describing complex natural processes in water objects. The proposed method of assessment and adjustment of the accumulation factor of ¹³⁷Cs in silty bottom sediments is quite rough, as it depends on the number of conservative assumptions of the two-chamber model and some intuitive assumptions in estimation of typical values of migration parameters.

One should be cautious while using the method for practical assessments of activity of ^{137}Cs in water on the base of observed activity in bottom sediments. Indeed, in practice, sampling is usually carried out not far from riverbanks (or from shores of lakes). In these regions one can often find pebble and coarse-grained sand with different content of silt. If the sampling spots are chosen incorrectly and quantity of silt there does not represent adequately quantity of silt in the water object on the whole, one can obtain significant errors in assessment of ^{137}Cs activity in water. Therefore the proposed method should be used only for rough preliminary estimations. One should realize that observed values of ^{137}Cs activity in water could differ significantly from assessed with the use of this method.

The adjustment of the accumulation factor on the base of observed concentration of suspended matter in water is necessary to increase accuracy of the assessment. In case of following the mentioned recommendations, the method of preliminary assessment of ^{137}Cs activity in water is adequate. Thus its use in number of cases can be justified.

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